

## REMARKS

Claims 1-9, 11 and 13-15 are pending in the present application. By this amendment, Applicants have amended claims 1, 2 and 14, cancelled claim 10, and added new claim 15. Applicants respectfully submit that the present application is in condition for allowance based on the discussion which follows.

Claims 1-3, 5-7, 9-11 and 13-14 were rejected under 35 U.S.C. § 103(a) as being unpatentable over Grosch, et al. (DE 19623611, of record) (hereinafter “Grosch”), and in view of Hefele, et al. (WO 98/37965) (hereinafter “Hefele”), and further in view of Sepulveda, et al. (US 4,613,427, of record) (hereinafter “Sepulveda”).

In order to further highlight novel features and elements of the present method over the prior art, by this amendment, Applicants have amended claims 1 and 14, by incorporating subject matter from previous claims 2 and 10. Accordingly, the amendments to claims 1 and 14 do not constitute new matter. Applicants respectfully submit that, upon review of the now pending claims, it will be clear that the present method presents novel and non-obvious subject matter over the prior art, which is further highlighted by the following discussion.

The present invention is directed to providing an epoxidation catalyst based on titanium zeolite in the form of extruded granules which is:

- easy to separate from the epoxidation reaction medium,
- have a good mechanical strength, and
- have a high catalytic activity and a high selectivity,

(see, present specification, ¶ [0004].

These problems are solved by the choice of an extrusion process in which a limited amount of a silicon derivative binder (more than 5% and less than 20%), selected from silicon derivatives, is used, and by using a pore-forming compound in an amount of 5 to 35%.

The effect of the amount of binder and of the addition of a pore-forming compound is illustrated by the test results provided with the Declaration Under 37 C.F.R. § 1.132 of Michel Strebelle, dated December 19, 2001, originally submitted in the parent application to this one, Application No. 09/555,149, and subsequently submitted in this application with the response filed August 29, 2008.

The table summarizing the test results is reproduced below and the tests have been numbered.

Test Number	Amount of SiO <sub>2</sub> after calcination (g/100g of TS-1)	Amount of melamine before calcination (g/100g of TS-1)	Weight loss through attrition (%)	Kinetic constant related to H <sub>2</sub> O <sub>2</sub> (k. 10 <sup>3</sup> min <sup>-1</sup> )
1	6	0	0.8	10
2	6	10	1.0	17
3	11	0	0.2	12
4	11	10	0.4	17
5	20	10	0.4	10
6	20	20	0.5	14

These results show that a good compromise must be found between mechanical strength and catalytic activity, such compromise being linked to an amount of binder of more than 5% and less than 20% by weight of zeolite.

Indeed, on the one hand, using lower amounts of binder leads to a decreased mechanical strength. This is illustrated by the following tests:

– tests 1 and 3 (no pore-forming substance) show that the weight loss through attrition increases from 0.2 to 0.8% when 6% of binder is used rather than 11%, and

– tests 2, 4 and 5 (10% of pore-forming agent) show that the weight loss through attrition increases from 0.4 to 1% when 6% of binder is used, rather than 11 of 20%.

These results show an inferior limit above 5% of binder, as below such limit, weight loss through attrition will become higher than 1%.

On the other hand, using higher amounts of binder leads to a drastic decrease of the catalyst activity. This is illustrated by the tests 4 and 5 (20% binder, 10% pore-forming agent), which show identical mechanical strength, but a decrease of the kinetic constant related to  $\text{H}_2\text{O}_2$  from  $17.10^3$  to  $10.10^3 \text{ min}^{-1}$ .

The binder will be converted into a material forming a matrix.

These results also show that another important characteristic of the present invention is the use of at least one pore-forming substance in an amount from 5 to 35% by weight of zeolite. Such pore-forming substances are eliminated during the calcination step (2). Their effect is to increase the porosity of the resulting catalyst. But surprisingly, their addition also leads to an improvement of the catalyst activity. As can be seen from the table above, the addition of a pore-forming substance in an amount of 10 or 20% by weight of zeolite leads to a clear increase of the kinetic constant related to  $\text{H}_2\text{O}_2$  (measurement of the catalyst activity), while the mechanical strength of the catalyst is maintained or only slightly decreased. This can be seen from the following tests:

– tests 1 and 2 (6% binder) show that the addition of melamine as pore-forming substance in an amount of 10% increases the kinetic constant related to  $\text{H}_2\text{O}_2$  from  $10.10^3$  to  $17.10^3 \text{ min}^{-1}$ , while the weight loss through attrition stays at a maximal value of 1%,

– tests 3 and 4 (11% binder) show an improvement from  $12.10^3$  to  $17.10^3 \text{ min}^{-1}$ , while the weight loss through attrition is only 0.4%, and

– tests 5 and 6 (20% binder) show an improvement from  $10.10^3$  to  $14.10^3 \text{ min}^{-1}$ , but which is not sufficient to compensate the decrease arising from the too high amount of binder compared to the claimed amount.

Applicants respectfully submit that it will be clear now that the present method is distinguishable over the prior art as further discussed in the following remarks regarding the respective prior art references cited in the 35 U.S.C. § 103(a) rejection.

Grosh discloses a process for the production of epoxides using an oxidation catalyst prepared by blending titanium silicalite zeolite powder, water, a binder (such as ammonium-stabilized silica sol) and a plasticizer (such as methylcellulose) and further extruding, drying and calcining the mixture. According to Grosh, the binder is added in an amount of 5% by weight of the zeolite (12.5% of a 40% solution) (see, Grosh, Example 1).

Grosh is silent about the use of a pore-forming agent, especially in an amount from 5 to 35% by weight of zeolite. Grosh is also silent about the use of the binder in an amount of more than 5% and less than 20%.

The effect of these differences is a better mechanical strength and a better catalytic activity compared to catalysts having a smaller amount of binder and no pore-forming compound.

Hefele discloses a process for preparing phthalic anhydride by catalytic gas-phase oxidation, using a coated catalyst comprising an inert, nonporous support material on which a catalytically active composition comprising titanium dioxide and vanadium pentoxide is applied in layer form (see, Hefele, claim 1). According to Hefele, the titanium dioxide and the vanadium pentoxide layer can be formed by applying the catalytically active composition or the precursor compound thereof on the support:

- by spraying a mix,
- by the application of a paste-like mass, or
- by the application of a powder,

comprising the catalytically active components of their precursors (see, Hefele, column 6, lines 8-30). Still, according to Hefele, if the coating is conducted by applying a paste-like mass, this paste-like composition may comprise auxiliaries, such as binders and pore formers (see, Hefele, column 6, lines 51-53).

Hefele does not relate to epoxidation catalysts. Hefele does not even refer to catalysts prepared by extrusion, nor to catalysts comprising titanium zeolites ( $TiO_2$  cannot be compared to titanium zeolites, which are specific compounds). One of ordinary skill in the art would, thus, not combine the teachings of Grosh and Hefele, as both documents do not relate to the same field. This combination of prior art documents is, thus, a hindsight reconstruction.

Furthermore, even if one of ordinary skill in the art combines both documents, they would not obtain the present invention. Hefele is silent about the amount of pore-forming agent and of binder to be used. According to Hefele, the type and amount of the auxiliaries is advantageously optimized in a preliminary experiment. But nothing in Hefele teaches to one of ordinary skill in the art that, in the field of expoxidation catalysts, especially epoxidation catalysts comprising titanium zeolites and prepared by extrusion, surprising effects can be obtained by using both a binder and a pore-forming agent in the specific amounts of the invention. Especially, nothing in Hefele, even combined with Grosh, teaches one of ordinary skill in the art that a compromise must be found between the mechanical strength and the activity of the catalyst, said compromise being linked to the use of both a binder selected from silicon derivatives and a pore-forming compound, and further linked to the amount of these products, especially the amount of binder.

Sepulveda discloses a process for the hydrodemetallization and hydroconversion of heavy hydrocarbon charge using a natural catalyst comprising a clay (see, Sepulveda, claim 1). According to Sepulveda, the catalyst is formed by extruding a mixture comprising the treated clay, water and a pore-forming substance in an amount of 5 to 40% by weight of dry clay.

First, Sepulveda does not relate to epoxidation catalysts, nor to catalysts comprising titanium zeolites. The skilled person would, thus, not combine the teaching of Sepulveda with the teachings of Grosh and/or Hefele. This combination of prior art documents is a hindsight reconstruction.

Second, Sepulveda is silent about the use of a binder, especially selected from silicon derivatives, in particular, in an amount of more than 5% and less than 20% by weight of zeolite.

Thus, even if the teaching of Sepulveda is combined with the teaching of Grosh and Hefele, one of ordinary skill in the art would not obtain the features of the present invention.

Based on the foregoing, Applicants respectfully submit that claims 1-9, 11 and 13-14 are not obvious in view of the cited prior art.

Further, in the Office Action, claims 1-14 were previously rejected on the ground of non-statutory obviousness-type double patenting over claims 1-13 of U.S. Patent No. 6,699,812 (the '812 Patent), in view of Grosch. With this amendment, Applicants have submitted a terminal disclaimer disclaiming the patent term of a patent issuing on the present application, which would extend beyond that of the '812 Patent, thereby obviating the double patenting rejection.

Finally, by this amendment, Applicants have reintroduced the subject matter of previously cancelled claim 12 as new claim 15. Applicants submit that new claim 15, dependent from claim 1 and based on original claim 12, is clear of the prior art for at least the reasons discussed above with regard to claim 1.

In light of the amendments and arguments provided herewith, Applicants submit that the present application overcomes all prior rejections and objections, and has been placed in condition for allowance. Such action is respectfully requested.

Respectfully submitted,

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